Sequential Extraction Methods: Applications in Identifying Geochemical Associations of Radiological Contaminants and in Developing NIST Natural Matrix Standards

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An Irish Sea standard sediment (IAES-135) was analyzed by sequential chemical extractions in order to determine the fractionation of actinide elements Am, Pu, and U. The sequential extraction methodology was evaluated and the potential artifacts: analyte readsorption; nonselectivity; and incomplete dissolution of geochemical target phases were investigated. The methods applied were derived from the NIST Specification Workshop, held June 13-15, 1995. The technique, as is applies to the development of NIST natural matrix standards which are certified by geochemical fraction, will be addressed.

The application of an interfacial rinse disodium ethylenediamine tetraacetic acid (EDTA) was investigated to determine the viability of this chelating agent in lessening analyte readsorption prior to the separation of aqueous and solid phases during sequential extraction procedures. Although the interfacial rinse was effective in alleviating readsorption to some extent, stable element analyses of the leachates from these experiments showed that the selectivity of the method was compromised. However, the combination of readsorption experiments and stable element analyses proved effective in identifying probable geochemical carrying phases for Am, Pu, and U, in this marine sediment. The extraction profiles of Am and Pu were found to show good correlation with that of Ca and Mn, indicating that these elements may be associated with a carbonate mineral phase and oxyhydroxides of Mn. Based on these same analyses natural uranium appears to be associated with the most refractory mineral phases of the sample. While Am showed no refractory characteristics, the extraction of profiles for Pu showed a strong signal in the residual fraction. Isotopic ratios of Pu-238:Pu-239/240 in this fraction suggest geochemical cycling of Pu to a more refractory phase over time.